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Title: Enhanced optical properties of in situ passivated near-surface Al<sub>x</sub>Ga<sub>1-x</sub>As/GaAs quantum wells

Year: 1996

Version: Final published version

**Please cite the original version:**

Lipsanen, Harri & Sopanen, M. & Taskinen, M. & Tulkki, J. & Ahopelto, J. 1996. Enhanced optical properties of in situ passivated near-surface Al<sub>x</sub>Ga<sub>1-x</sub>As/GaAs quantum wells. Applied Physics Letters. Volume 68, Issue 16. P. 2216-2218. ISSN 0003-6951 (printed). DOI: 10.1063/1.115863.

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## Enhanced optical properties of in situ passivated near-surface $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum wells

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Citation: *Applied Physics Letters* **68**, 2216 (1996); doi: 10.1063/1.115863

View online: <http://dx.doi.org/10.1063/1.115863>

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# Enhanced optical properties of *in situ* passivated near-surface $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum wells

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(Received 25 October 1995; accepted for publication 13 February 1996)

An epitaxial method for *in situ* passivation of epitaxial  $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  surfaces is reported. The deposition of an ultrathin InP layer (about one monolayer) on the surface of  $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  structures by metalorganic vapor phase epitaxy results in drastically reduced surface recombination. The effect is studied by low-temperature photoluminescence of near-surface  $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}/\text{GaAs}$  quantum wells where the top barrier thickness is varied from 0 to 50 nm. At the thicknesses of  $\leq 5$  nm, the intensity from passivated samples is more than four orders of magnitude larger than that obtained from unpassivated structures. For a passivated surface quantum well where InP is deposited directly onto the GaAs quantum well, we observe a blueshift of 15 meV and an intensity reduction of only a factor of 10 as compared to the luminescence from a quantum well placed at a depth of 50 nm from the surface. © 1996 American Institute of Physics. [S0003-6951(96)04316-1]

Surface states have a detrimental effect on the optical and electronic properties of modern semiconductor devices that are located in close proximity to the surface. Large surface recombination velocity and Fermi level pinning are consequences of these states. Various surface treatment techniques have been proposed to passivate the surface. Sulfide solution passivation of a GaAs surface has been studied intensively in recent years.<sup>1</sup> A monolayer of sulfur atoms terminates the dangling bonds and effectively reduces the surface-state density as also shown by *ab initio* calculations.<sup>2</sup> The sulfur passivation and many other treatments, such as ion-gun hydrogenation,<sup>3</sup> nitride deposition,<sup>4</sup> and evaporated overlayers<sup>5</sup> are generally performed on etched or cleaned surfaces. However, the most efficient way to suppress the surface state density of epitaxial layers is an *in situ* surface passivation. Two promising techniques for the *in situ* GaAs surface passivation have been demonstrated. Stabilization of GaAs surfaces by phosphorus is based on the As–P exchange reaction that forms a thin GaP layer on the surface.<sup>6</sup> The method has been shown to increase the photoluminescence (PL) intensity by a factor of 4. Wada and Wada reported that the deposition of a thin InP layer on an *n*-type GaAs wafer results in tenfold enhancement in room temperature PL intensity partly due to reduced band bending.<sup>7</sup> The estimated layer thickness deposited at 550 °C was 1 nm. These two approaches were developed for GaAs, but no practical *in situ* passivation of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  has been reported.

We report an *in situ* passivation technique based on a very thin epitaxial InP layer. The technique has been used for epitaxial GaAs and also  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  surfaces. The surface recombination in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  is an even more severe problem than in GaAs due to oxidation. InP and related compounds are known to have a considerably smaller surface recombination velocity than GaAs or  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ .<sup>8</sup> It has also been suggested that the band gap modulation due to

strain in InP on GaAs has a contribution to the reduced surface recombination.<sup>7</sup> Since InP has a 3.8% larger lattice constant than GaAs, the compressively strained two-dimensional InP layer can be only about 1–2 monolayers (ML) thick without generation of self-organized islands<sup>9</sup> or dislocations at typical growth temperatures. We have chosen a passivation layer thickness of 1 ML (about 0.3 nm) in this letter. Near-surface quantum wells (QWs) are very sensitive probes to surface states.<sup>10–13</sup> The band bending in these structures does not have such a major impact on PL intensity as it does in bulk semiconductors. Nevertheless, at low temperatures, the strong photovoltage effect due to pump light tends to flatten the bands, which suppresses the drift of minority carriers to nonradiative surface states. The surface recombination in both GaAs and  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  due to the Fermi level pinning can be reduced by adding a larger band gap layer, e.g., an  $\text{Al}_z\text{Ga}_{1-z}\text{As}$  layer with  $z > x$  to the surface.<sup>14</sup> However, this approach does not affect the surface state density.

The PL intensity decreases rapidly when the QW is closer than about 10 nm from the surface. Moison *et al.*<sup>10</sup> fabricated near-surface  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}/\text{GaAs}$  QWs and observed a redshift of the luminescence peak as the cap layer thickness was reduced, instead of the blueshift expected. They explained this as being due to an interaction of the QW states and the surface states. A significant blueshift of 20 meV was observed by Dreybrodt *et al.* for  $\text{Ga}_{0.8}\text{In}_{0.2}\text{As}/\text{GaAs}$  surface QWs.<sup>11</sup> We show, for the first time, that a very high PL intensity and a blueshift of 15 meV is obtained from an  $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  surface QW with InP passivation.

The samples were grown by metalorganic vapor phase epitaxy (MOVPE) at atmospheric pressure using trimethylgallium (TMGa), trimethylindium (TMIn), trimethylaluminum (TMAI), tertiarybutylarsine (TBAs), and tertiarybutylphosphine (TBP). The structures consist of a 20 nm thick GaAs buffer layer, a 200 nm thick  $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$  layer, a 5 nm thick GaAs QW, and a  $d=0$  to 50 nm thick  $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$  top barrier layer. The nominally undoped lay-

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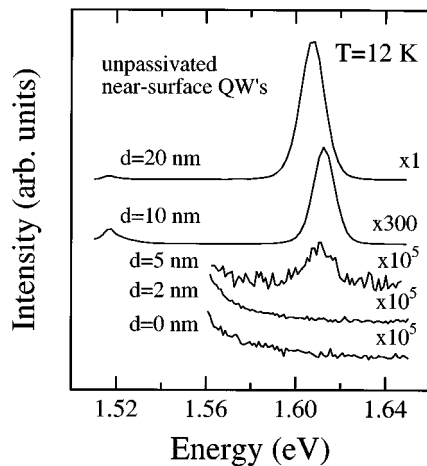


FIG. 1. Low-temperature PL spectra of unpassivated near-surface  $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}/\text{GaAs}$  quantum wells for various top barrier thicknesses  $d$ . The intensity decreases rapidly for  $d < 10$  nm.

ers were grown on semi-insulating  $(100) \pm 0.5^\circ$  GaAs substrates at  $670^\circ\text{C}$ . The estimated  $p$ -type carrier concentration in AlGaAs is at a high- $10^{16} \text{ cm}^{-3}$  level. Two sets of samples were fabricated. After the growth of the QW structure, the passivation was performed by switching the TBAs flow to the TBP flow at  $670^\circ\text{C}$ . After a 2 s pause, 1 ML of InP was deposited in 0.7 s using the V/III ratio of 100. The surface was protected with a TBP flow down to  $400^\circ\text{C}$ . The unpassivated reference samples were cooled to room temperature immediately after the QW growth using a protective TBAs flow down to  $400^\circ\text{C}$ . The as-grown samples were kept in nitrogen ambient prior to transfer in air to optical measurements.

The PL measurements were carried out in a closed-cycle helium cryostat at 12 K. A 488 nm line from an argon-ion laser was used for pumping. The spectra were recorded with an 0.5 m monochromator and a photomultiplier tube. The intensity from the passivated samples was found to depend linearly on the pump intensity over three orders of magnitude. Due to the very low intensity from the unpassivated samples with  $d < 10$  nm, a pump intensity of  $30 \text{ W/cm}^2$  was chosen for the measurements.

Figure 1 shows the PL curves of the QW samples without passivation. The lowest QW transition associated with heavy holes is seen at about 1.60 eV. The weak signal at 1.52 eV is due to the near-band edge GaAs transitions from the substrate or from the buffer layer. A rapid decrease in intensity is observed as the top barrier thickness  $d$  is lowered to 10 nm. A very weak peak at 1.610 eV is observed for  $d = 5$  nm but no signal is observed from the QW for  $d = 0$  or 2 nm. The lowest two spectra are dominated by the slope from the near-band edge GaAs transitions. The results observed are in good agreement with previous reports.<sup>10,12</sup> The PL spectra of samples passivated by InP are shown in Fig. 2. The intensity decreases only by about an order of magnitude as the top barrier thickness is reduced from  $d = 20$  nm to  $d = 0 - 5$  nm. Actually, the intensity increases when the passivation is performed directly on the GaAs well, i.e.,  $d = 0$  nm instead of a thin  $d = 2 - 5$  nm AlGaAs layer. At the same

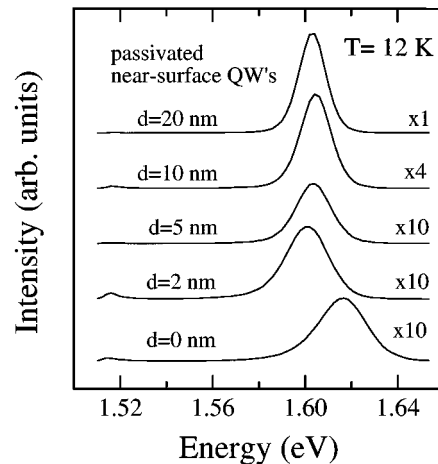


FIG. 2. PL spectra of near-surface QW's passivated *in situ* by a 1 ML thick InP layer as a function of top barrier thickness  $d$ . A very high PL intensity is also obtained for the surface QW.

time, the QW peak shifts to blue by about 15 meV. We attribute the increased intensity due to lower surface state density in GaAs than in AlGaAs and the blueshift due to a high-quality surface barrier, where the surface states do not dominate as proposed in Ref. 10. The measured blueshift is about half of the theoretical value of 33 meV calculated using previously reported assumptions for the surface barrier.<sup>11</sup> The blueshift was calculated from single conduction band and four valence band (Luttinger-Kohn) models by including the pertinent strain potentials according to the theory of Pikus and Bir.<sup>15</sup>

The integrated intensities from the passivated and reference samples are compared in Fig. 3. The intensities were measured after a few hours from the growth and again about 5 days later with the samples stored in ambient air. The oxidation had no noticeable effect on the intensity of the passivated samples, whereas the intensity from the reference samples with  $d \leq 10$  nm decreased by about an order of magnitude when compared to as-grown samples. The passivated

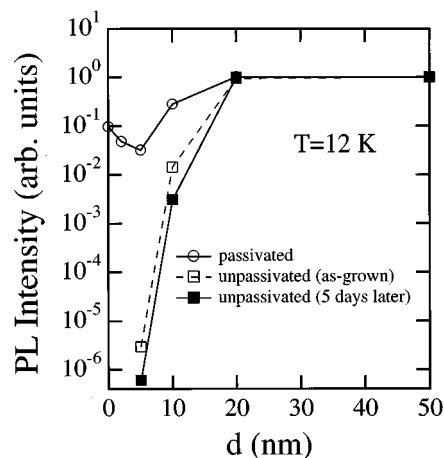


FIG. 3. Integrated PL intensity of passivated (open circles) and unpassivated QW's after a few hours from the growth (open squares) and after about five days (filled squares) as a function of top barrier thickness  $d$ . The passivated samples showed no degradation after 5 days.

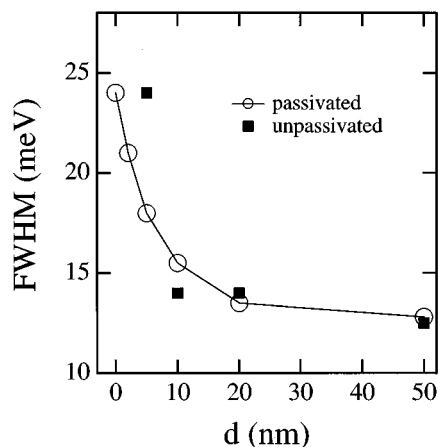


FIG. 4. FWHM of the passivated (open circles) and unpassivated (filled squares) QWs as a function of top barrier thickness  $d$ . The FWHM increases from 13 to 24 meV for a deep QW and a surface QW, respectively.

samples show as much as  $10^5$  times larger intensities than the reference samples when the QW is 0–5 nm from the surface. For the surface QW, the ratio can be even larger ( $>10^6$ ) if the noise level is taken as the reference signal. The very large intensity ratio is distinct evidence of the effective surface passivation. The PL intensity from the passivated surface QW after 1 month from the growth was only 25% lower than that of the as-grown sample. This shows the good long-term stability of the passivation. Another reference QW sample was grown consisting of a 5 nm top  $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$  barrier and a 2 nm thick GaAs surface layer, i.e., the QW is at the depth of 7 nm from the surface. The PL intensity from it coincided closely with the interpolated intensity from an unpassivated sample with  $d=7$  nm. Therefore, unpassivated AlGaAs and GaAs surfaces show similar characteristics as probed by PL from near-surface QWs.

The full-width at half-maximum (FWHM) of the PL peaks is shown in Fig. 4. The FWHM of the passivated samples increases by a factor of 2 from 13 to 24 meV as  $d$  is decreased from 50 to 0 nm. This broadening is smaller than the previously reported broadening from 10 to 40 meV for 5 nm thick  $\text{Ga}_{0.8}\text{In}_{0.2}\text{As}/\text{GaAs}$  near-surface QWs.<sup>11</sup> We studied

the effect of a thicker 2 ML InP passivation layer on PL of the surface QW. The FWHM was 35 meV and the intensity was lower. The narrow half-width obtained in this work with optimized 1 ML InP passivation is assumed to be due to a smooth surface and a thin surface oxide layer and indicates the high quality of the structure.

In conclusion, we have demonstrated an efficient method to passivate the surface of epitaxial GaAs,  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ , and  $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  heterostructures by *in situ* deposition of InP on the surface. One monolayer of InP was found to be sufficient for the passivation. The passivation was studied by photoluminescence of near-surface  $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}/\text{GaAs}$  quantum wells. The luminescence intensity in passivated samples increased by about five orders of magnitude for quantum wells located at less than 5 nm from the surface as compared to unpassivated samples. Furthermore, we observed a blueshift of 15 meV for a passivated surface quantum well.

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